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LONG-LIVED COBALT ISOTOPES OBSERVED IN FALLOUT FROM THE NAVAJO DETONATION OF OPERATION REDWING

Research and Development Technical Report USNRDL-TR-215
NS 081-001

26 March 1958

by

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J.L. Mackin
D. MacDonald
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U.S. NAVAL RADIOLOGICAL DEFENSE LABORATORY

SAN FRANCISCO 24 CALIFORNIA

RESTRICTED DATA
ATOMIC ENERGY ACT--1954

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Effects of Atomic Weapons

Technical Objective
AW-7

Analytical and Standards Branch
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ABSTRACT

Analyses of three samples of fallout from the Navajo detonation of Operation REDWING revealed the presence of the induced radioisotopes Co^{57}, Co^{58} and Co^{60}. The amounts of these radioisotopes present in the samples would not constitute an internal hazard compared to the amounts of Sr^{90} present in the samples. However, the Co^{57}, Co^{58} and Co^{60} values obtained indicate that these isotopes would contribute appreciably to the external gamma radiation from fallout fields represented by the samples. The product/fission ratios of the individual nuclides were obtained.

NONTECHNICAL SUMMARY

The Problem

It is possible for radioactivity to be induced in environmental materials by radiation from a nuclear detonation. If such formation occurs to an appreciable extent, the radioisotopes thus created could be in the fallout resulting from the detonation and would increase the radiological hazard beyond that which would derive from the fission products of the weapon itself.

Evidence was found that cobalt-60 had been induced during nuclear weapon test detonations. As this radioisotope is long-lived and emits gamma radiation of appreciable energy and therefore could be significant in fallout hazards, a search was made, in fallout samples, for cobalt radioisotopes produced during Operation REDWING, Shot Navajo, and quantitative measurements were made of those found.

Findings

Three induced radioisotopes of cobalt were found, Co^{57}, Co^{58} and Co^{60}. Compared to the strontium-90 also found, these radioisotopes would not constitute an internal hazard if the fallout were to be inhaled or ingested. However, they would contribute significantly to the external hazard from the gamma radiation emitted by fallout fields represented by the samples.

ADMINISTRATIVE INFORMATION

The work reported herein was sponsored by the Bureau of Ships of the Navy under Technical Objective AW-7, NS 081-001 as Program 25; Problem 1 during Fiscal Year 1958. This work is described in this Laboratory's Proposed Technical Program for Fiscal Year 1958 dated May 1957.

REPORT OF INVESTIGATION

INTRODUCTION

It has been assumed generally that the long-term radiological hazards due to fallout from a nuclear detonation depend primarily upon the long-lived fission products. However, other radioactive products which are not derived from fission reactions may be present in fallout and contribute to the hazard. These latter products would be created by neutron activation of elements in the immediate vicinity of the detonation. The presence of one such product, Co^{60}, has been indicated by analyses of fallout material* and of biological specimens recovered from fallout areas.[1]

The present investigation, carried out to determine the amounts of Co^{60} in fallout samples from the Navajo detonation of the 1956 test series, revealed the existence of two additional cobalt nuclides, Co^{57} and Co^{58}. The amounts of these nuclides were determined. To permit assessment of the relative internal hazards from the cobalt isotopes, determinations of Sr^{90} were made on the same samples. From these data the cobalt-60, -58, -57/strontium-90 ratios were derived. Since all three cobalt isotopes emit gamma radiation, they would also contribute to an external hazard. This was estimated by summing the photon energy release per unit time from the observed amounts of radiocobalt and comparing it with similar values calculated for the associated fission products.

The total number of fission events represented by each sample was determined. Product-to-fission ratios for the radiocobalt isotopes in each sample were computed.

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* Private communication, D. Sam of this laboratory.

EXPERIMENTAL

Radiochemical analyses were carried out on three samples of fallout which had been collected in large trays at approximately 25 miles NNW of ground zero (sample designated YAG-39-35 Na) and at about 6 miles SE of ground zero (samples designated YFNB-13E-59 Na and YFNB-13E-60 Na).

The total number of fissions in each sample was determined by counting gross fission-product activity with a special low-geometry gamma scintillation counter. The counter was previously calibrated, by radiochemical methods, in terms of counts per minute per fission as a function of time.[2,3]

After the fission evaluations, the material composing each sample was removed from the collector trays and was brought into solution by successive treatment with fuming HNO_3, $HClO_4$ and HF. Each sample was then taken to a volume of 100 ml with 6N HCl. Aliquots were removed from the solutions for cobalt and strontium analyses.

The strontium analyses were carried out through modification of an existing method.[4] Step-wise, the procedure was as follows:

1. A 25-ml aliquot of the original solution was placed in a 50-ml centrifuge tube and reduced to 5 ml by heating.

2. Five hundred λ of carrier-free Sr^{85} were added for yield determination.

3. Two ml of strontium carrier ($\sim$ 6.00 mg Sr^{++}/ml) were added. Following this, 30 ml of fuming HNO_3 were added, the cone was placed in an ice bath for several minutes, and the aliquot was stirred, removed and centrifuged.

4. The supernatant was decanted and the precipitate dissolved in 10 ml of water. Ten drops of Fe^{+++} solution ($\sim$ 10 mg Fe^{+++}/ml) were added and the solution was heated to boiling. Iron was precipitated with carbonate-free NH_4OH and the solution was centrifuged. The time of separation of Y from Sr was noted, and the supernatant was transferred to a clean centrifuge cone.

5. After addition of 2 ml of conc NH_4OH to the clear supernatant, the solution was heated to boiling and 5 ml of saturated $(NH_4)_2C_2O_4$ solution was added drop-wise with stirring. The resulting precipitate was filtered onto a No. 42 Whatman filter paper, washed three times with 5 ml of water, three times with 5 ml of ethanol and three times with 5 ml of ether.

6. The precipitate was dried in an oven at 100°C for 20 min, cooled and weighed as $SrC_2O_4 \cdot H_2O$. It was then placed on a thick brass planchet and beta counted.

7. Following beta counting, the sample was transferred to a test tube. The filter paper was wet-ashed and the Sr brought into solution with 1 ml of water.

Prior to final solution of the separated Sr (Step 7), the growth of Y^{90} was followed. These measurements were used to compute the amount of Sr^{90} at time of separation (Step 4). After application of the usual corrections, the number of Sr^{90} atoms present at zero time was determined, using a half-life value of 27.7 ± 0.4 years.[5]

The amount of Sr^{85} present in the final solution was measured with a gamma ray spectrometer. This value was compared to the amount of Sr^{85} added and the yield derived.

Analyses for cobalt were carried out through modification of a published method.[4] Since Mn^{54} was known to be present, manganese carrier was added.[6] Step-wise, the analytical procedure was as follows:

1. Two ml of standardized cobalt carrier (~ 10 mg Co/ml) and 1 ml of Mn carrier (~ 10 mg Mn/ml) were added to a 25-ml aliquot of original solution.

2. The solution was heated to dryness and 0.5 ml conc HCl and 3 ml of water were added.

3. One ml of 5M NH_4Cl solution was added. The resulting solution was made alkaline with 15M NH_4OH and 10 drops excess NH_4OH added. After this the solution was centrifuged and the precipitate discarded.

4. The supernatant was saturated with H_2S and then centrifuged. Two drops of 15M NH_4OH were added and H_2S was passed through the supernatant. After this the supernatant was discarded and the precipitate was washed with 3 ml of water.

5. Three ml of a solution of equal parts of saturated Na_2SO_4 and 2M $NaHSO_4$, by volume, were added. The resulting solution was stirred for 2 min, and then centrifuged. The supernatant was discarded and the precipitate was washed with 3 ml of water.

6. One-half ml of conc HCl, 1 drop of H_2O_2 and heat were used to bring the precipitate into solution. Two ml of glacial acetic acid were added and the solution diluted to 25 ml with water.

7. Six ml of 3M acetic acid saturated with KNO_2 were added and the mixture was cooled in an ice bath for ten min. The solution was centrifuged, the supernatant removed and the precipitate washed with 30 ml of 1.5M acetic acid and 30 ml of a 50% ethyl alcohol-water solution.

8. Three ml of conc HCl were used to dissolve the precipitate. The solution was heated to boiling and Cs and Ba carriers added.

9. The solution was diluted to 25 ml with water. Ten M KOH were added to precipitate $Co(OH)_2$. The mixture was centrifuged, the supernatant discarded and the precipitate washed with 5 ml of water.

10. The $Co(OH)_2$ was dissolved with 3 ml of 6M acetic acid and Cr and Zr holdback carriers were added. Water was used to dilute to 25 ml and 6 ml of 3M acetic acid saturated with KNO_2 were added to precipitate $K_3Co(NO_2)_6$.

11. The precipitate was filtered onto a Whatman 42 filter paper and washed twice with 30 ml of 1.5M acetic acid and twice with 30 ml of 50% ethyl alcohol-water solution.

12. The cobaltinitrite precipitate was placed in an oven and dried for 20 min at 100°C. It was cooled and weighed to determine yield.

The cobalt yields were dissolved and analyzed for gamma activities, using a single-channel gamma pulse-height analyzer. The presence of Co^{60}, Co^{57} and Co^{58} was established by identification of their respective photopeaks by comparison with standards of known energy.

The disintegration rate of each cobalt isotope was obtained by analysis of photopeak areas. After determination of the total pulse height distribution, a standard of Co^{60} was prepared to equal the magnitude of the Co^{60} photopeaks of the isolated cobalt. This Co^{60} pulse height distribution was subtracted from the total distribution. This subtraction of the Co^{60} Compton continuum permitted determination of the Co^{57} and Co^{58}. Their disintegration rates were computed from experimentally determined gamma ray efficiencies and consideration of their respective decay schemes (relative photon abundance, internal conversion, etc.).[7] Figure 1 shows the pulse height distribution of the separated cobalt and the spectrum of the Co^{60} standard. The 0.84-Mev Mn^{54} photopeak is shown as an energy reference.

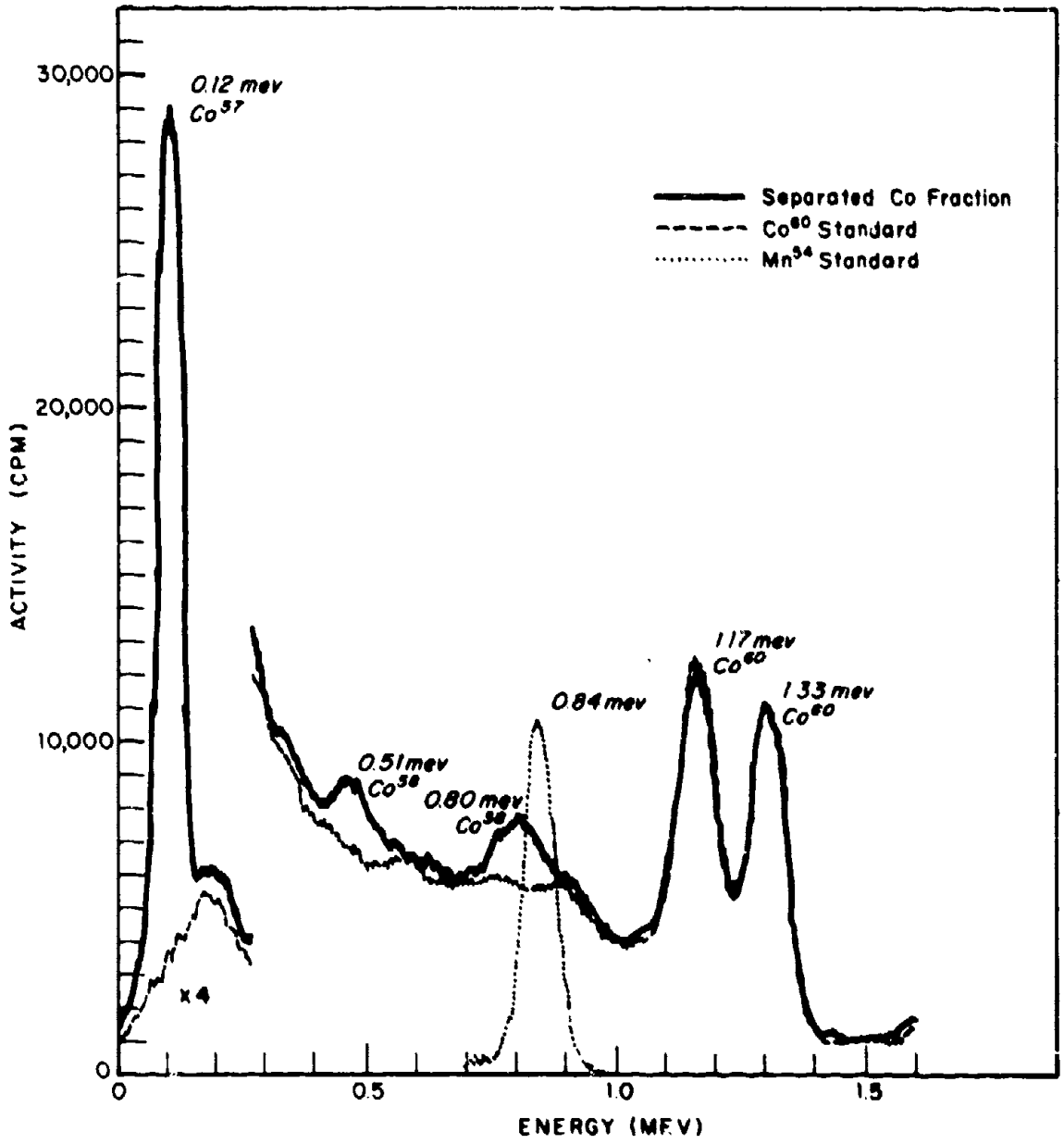

Fig. 1 Comparison of Pulse Height Distribution of Separated Cobalt Sample With Distributions of Reference Standards

RESULTS

The quantities of the isotopes Co^{57}, Co^{58} and Co^{60} present in each sample at zero time and the quantities of Sr^{90} present in each sample at zero time are given in Table 1. This table also includes the fission numbers determined for each sample. The error associated with each value in Table 1 is the determinate error involved in the measurements; these include pipetting error, counting error, errors in chemical yields, etc.

TABLE 1

Atoms Present at Zero Time and Total Fissions in Each Sample

Sample[a]	Co^{60}	Co^{57}	Co^{58}	Sr^{90}	Fissions
A	$3.72\pm1.14\times10^{11}$	$5.83\pm1.87\times10^{10}$	$3.13\pm2.25\times10^{10}$	$1.15\pm0.08\times10^{12}$	$4.13\pm0.41\times10^{13}$
B	$1.25\pm0.49\times10^{12}$	$1.96\pm0.69\times10^{11}$	$1.05\pm0.76\times10^{11}$	$5.17\pm0.38\times10^{12}$	$1.44\pm0.14\times10^{14}$
C	$1.18\pm0.36\times10^{12}$	$1.85\pm0.65\times10^{11}$	$1.01\pm0.73\times10^{11}$	$4.62\pm0.34\times10^{12}$	$1.36\pm0.14\times10^{14}$

(a) Sample A, YAG-39-35 Na
Sample B, YFNB-13E-59 Na
Sample C, YFNB-13E-60 Na.

The data of Table 1 were used to compute the $Co^{57,58,60}/Sr^{90}$ ratios listed in Table 2. The ratio errors shown are the determinate errors associated with each measurement. The average errors are shown with the average ratio values obtained.

Product-to-fission ratios for the individual cobalt nuclides in each sample were derived from the data of Table 1. These ratios are presented in Table 3. Once again, determinate errors are included. Average errors are associated with the average product-to-fission values.

TABLE 2

Ratios of Abundance of Co Atoms to Sr^{90} Atoms at Zero Time

Sample	Co^{60}/Sr^{90}	Co^{57}/Sr^{90}	Co^{58}/Sr^{90}
A	0.323 ± 0.106	0.051 ± 0.017	0.027 ± 0.020
B	0.245 ± 0.102	0.038 ± 0.013	0.021 ± 0.015
C	0.255 ± 0.090	0.040 ± 0.013	0.022 ± 0.016
Av Values	0.274	0.043	0.023
Av Errors	± 0.100	± 0.014	± 0.017

TABLE 3

Product-to-Fission Ratios of the Cobalt Isotopes

Sample	Co^{60}/f	Co^{57}/f	Co^{58}/f
A	$9.01 \pm 2.91 \times 10^{-3}$	$1.41 \pm 0.47 \times 10^{-3}$	$7.58 \pm 5.50 \times 10^{-4}$
B	$8.68 \pm 2.80 \times 10^{-3}$	$1.36 \pm 0.45 \times 10^{-3}$	$7.29 \pm 5.32 \times 10^{-4}$
C	$8.68 \pm 2.80 \times 10^{-3}$	$1.36 \pm 0.45 \times 10^{-3}$	$7.43 \pm 5.42 \times 10^{-4}$
Av Values	8.79×10^{-3}	1.38×10^{-3}	7.43×10^{-4}
Av Errors	$\pm 2.84 \times 10^{-3}$	$\pm 0.46 \times 10^{-3}$	$\pm 5.41 \times 10^{-4}$

DISCUSSION AND CONCLUSIONS

The ratios shown in Table 2 indicate that significant amounts of radiocobalt were formed relative to Sr^{90}. The product-to-fission values of Table 3 show that there was no apparent radiocobalt fractionation between the two fallout positions sampled. Possible reactions for the formation of the cobalt nuclides from environmental nickel and cobalt are: $Co^{59}(n,\gamma)Co^{60}$, $Ni^{60}(n,\ )Co^{60}$, $Co^{59}(n,2n)Co^{58}$, $Ni^{58}(n,p)Co^{58}$ and $Ni^{58}(n,2n)Ni^{57} \xrightarrow{\beta+,\ K} Co^{57}$.

The biological hazards resulting from fallout may be placed into two categories:

1. the internal hazard caused by ingestion and inhalation of active material and

2. the external hazard caused by penetrating radiation, the source of which is external to the body.

If material, whose atom ratios were that found in Table 2, were ingested or inhaled, the hazard due to the presence of the radiocobalt would be negligible as compared to the Sr^{90}. This is evident when, among other factors, the biological half-lives (3.9×10^3 d for Sr^{90} vs 8.4 d for Co^{60})[8] and the ionizing radiation emitted (β for Sr^{90}, primarily γ for $Co^{57,58,60}$) are compared.

External hazard in fallout may be considered as that primarily due to gamma radiation. This hazard is a function of photon energy, scattering, absorption, etc. The net effect, however, of these factors for fallout situations (approximated by uniformly contaminated fields) is that the hazard is roughly proportional to energy release per unit time. Values of gamma energy released per second (μ_1) of fission products resulting from 10^4 fissions have been computed for times up to 1.2×10^3 y after time of fission.[9] The values so obtained may be compared over the same time period with the energy released per second (μ_2) by the quantities of radiocobalt found associated with 10^4 fissions. This ratio μ_2/μ_1 in percent is found to be 0.14 at 9.8 d, 31 at 1.20 y, 149 at 2.6 y, 211 at 5.6 y, and 26 at 25.7 y. These values represent the analyses of a limited number of samples from one detonation. It is evident, however, that radiocobalt produced in the ratios given in Table 2, would constitute a considerable portion of the external hazard of fallout, particularly at times greater than 1 year.

Approved by:

E. R. Tompkins

E. R. TOMPKINS
Head, Chemical Technology Division

For the Scientific Director

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Defense Threat Reduction Agency
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CPWS/TRC March 26,1999

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Ardith Jarrett

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